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Segmented Power Generator Modules of Bi_2Te_3 and ErAs:InGaAlAs Embedded with ErAs Nanoparticles

Gehong Zeng[1], Je-Hyeong Bahk[1], John E. Bowers[1], Hong Lu[2], Joshua M. O. Zide[2], Arthur C. Gossard[2], Rajeev Singh[3], Zhixi Bian[3], Ali Shakouri[3], Suzanne L. Singer[4], Woochul Kim[4], and Arun Majumdar[4]

[1]Department of Electrical and Computer Engineering, University of California, Santa Barbara, CA, 93106

[2]Materials Department, University of California, Santa Barbara, CA, 93106

[3]Electrical Engineering Department, University of California, Santa Cruz, CA, 95064

[4]Department of Mechanical Engineering, University of California, Berkeley, CA, 94720

ABSTRACT

We report the fabrication and characterization of segmented element power generator modules of 254 thermoelectric elements. The element is 1 mm × 1 mm in area, which consists of 300 μm thickness Bi_2Te_3 and 50 μm thickness ErAs:$(InGaAs)_{1-x}(InAlAs)_x$, so that each segment can work at different temperature ranges. Erbium arsenide metallic nanoparticles are incorporated to create scattering centers for middle and long wavelength phonons, provide charge carriers, and form local Schottky barriers for electron filtering. The thermoelectric properties of ErAs:InGaAlAs were characterized by variable temperature measurements of thermal conductivity, electrical conductivity and Seebeck coefficient from 300 K to 600 K. Generator modules of Bi_2Te_3 and ErAs:InGaAlAs segmented elements were fabricated and an output power over 5.5 W was measured. The performance of the thermoelectric generator modules can further be improved by improving the thermoelectric properties of the element material, and reducing the electrical and thermal parasitic losses.

INTRODUCTION

The performance of a thermoelectric generator module depends largely on the material's thermoelectric properties, which are often summarized with the figure of merit, $Z = \alpha^2\sigma/\kappa$, where α is Seebeck coefficient, σ is electrical conductivity and κ is thermal conductivity. Thermoelectric properties can be improved by introducing nanometer scale structure into materials: the power factor ($\alpha^2\sigma$) can be enhanced because of the quantum confinement effect;[1] thermal conductivity can be reduced due to the increase of phonon interface scattering;[2] and the Seebeck coefficient can be increased through thermionic emission.[3-6] Thermal conductivity reduction using superlattice heterostructures or incorporation of nanoparticles has been demonstrated[7-9], and most of the recent ZT improvements come

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14. ABSTRACT

We report the fabrication and characterization of segmented element power generator modules of 254 thermoelectric elements. The element is 1 mm ? 1 mm in area, which consists of 300 μm thickness Bi2Te3 and 50 μm thickness ErAs:(InGaAs)1-x(InAlAs)x, so that each segment can work at different temperature ranges. Erbium arsenide metallic nanoparticles are incorporated to create scattering centers for middle and long wavelength phonons, provide charge carriers, and form local Schottky barriers for electron filtering. The thermoelectric properties of ErAs:InGaAlAs were characterized by variable temperature measurements of thermal conductivity, electrical conductivity and Seebeck coefficient from 300 K to 600 K. Generator modules of Bi2Te3 and ErAs:InGaAlAs segmented elements were fabricated and an output power over 5.5 W was measured. The performance of the thermoelectric generator modules can further be improved by improving the thermoelectric properties of the element material, and reducing the electrical and thermal parasitic losses.

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mainly from the reduction of thermal conductivity. In our study, ErAs nanoparticles, a rocksalt semimetal nanostructure, were epitaxially incorporated into $(InGaAs)_{1-x}(InAlAs)_x$ using molecular beam epitaxy (MBE). The ErAs nanoparticles in the InGaAlAs can provide both charge carriers, and create phonon scattering centers. Experiments show that the ErAs nanoparticles form effective phonon scatting centers for middle and long wavelength phonons, and therefore the thermal conductivity can be reduced. When ErAs nanoparticles are incorporated into $(InGaAs)_{1-x}(InAlAs)_x$, Schottky barriers are formed at the interface between the particle and semiconductor. The Schottky barrier height varies with temperatures and can be adjusted by varying the InAlAs concentration, which can be optimized to act as an energy barrier for hot carriers. The Seebeck coefficient can therefore be enhanced through the electron filtering effects of these Schottky barriers.[10]

In addition to good thermoelectric properties of the elements, generator modules must have large temperature difference across its elements to get large output power and efficiency, which requires the optimization of the element material thermoelectric properties within the range of the temperature difference. Using segmented elements, it will become possible for the generator module to work at large temperature range with each element segment working in the temperature range, where its thermoelectric properties are optimized.

In this paper, we report our 254 segmented element generator modules of ErAs:$(InGaAs)_{1-x}(InAlAs)_x$ alloy material and bulk Bi_2Te_3. The generator modules were fabricated via pickup and place method, and flip-chip bonding technique. The output power density of 5.5 W was measured with heat source temperature at around 600 K.

MATERIAL CHARACTERIZATION

The role of incorporating ErAs nanoparticles is to reduce the thermal conductivity below that of InGaAs or InGaAlAs. It has been shown previously that incorporating ErAs nanoparticles 1-5 nm in diameter effectively reduces the thermal conductivity below InGaAs because ErAs nanoparticles effectively scatter long and middle wavelength phonons while atomic substitution in InGaAs alloy scatters short wavelength phonons. Thermal conductivity of $(InGaAs)_{0.8}(InAlAs)_{0.2}$ with 0.3% ErAs nanoparticles was measured using the 3ω method. [9]

Samples with epitaxial layers of 0.3% ErAs: $(InGaAs)_{0.8}(InAlAs)_{0.2}$ with 0.5 and 2 μm thickness, respectively were grown on semi-insulating InP substrates about 520 μm thick using MBE for the Seebeck coefficient and electrical conductivity measurements. The 2 μm epitaxial layer samples were processed for Seebeck coefficient measurements. The test chips were cut into 5×20 mm^2 in strip size, and coated with 300 nm SiN on both the top and bottom sides. The contact electrodes of Ni/GeAu metallization and barrier metal layers were made at the each end of the strip chip. The test setup consists of two copper bars embedded with heaters placed in a vacuum chamber, and the test sample was placed across the two separated bars, which were set at different temperatures. The temperature difference ΔT and output voltage V were measured directly and the Seebeck coefficient α was the slope of the linearly fitted V vs. ΔT, which is expressed as $\alpha = V/\Delta T$. Different Seebeck coefficient test samples from the same material wafer were measured separately at both University of California at Santa Barbara (UCSB) and University of California at Santa Cruz (UCSC) to ensure that measurements were carried out

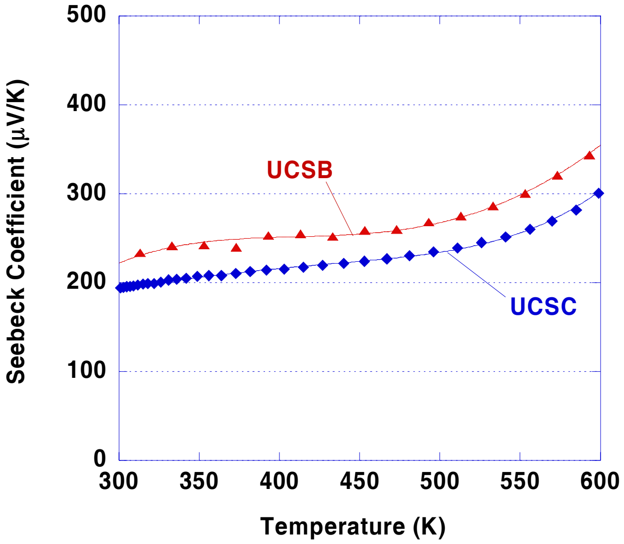

Figure 1. Seebeck coefficients of (InGaAs)0.8(InAlAs)0.2 with 0.3% ErAs nanoparticles vs temperatures from 300 K to 600 K. The measurements were separately carried out at UCSB and UCSC.

properly. The results from the two groups showed the similar trends of Seebeck coefficients versus temperatures from 300 K to 600 K, as shown in Figure 1.

Epitaxial layer samples 0.5 μm thick were used for van der Pauw measurements. The test samples were cut into 1 cm × 1 cm square chips. The sample was coated with 300 nm Si_3N_4 as a protection layer to prevent oxidation or diffusion of the material at high temperatures. Four contact metal electrodes of contact metallization of Ni/GeAu and barrier metal layers were deposited on the four corners. The measurements were carried out in vacuum around 1×10^{-3} Torr. The test chips from the same wafer samples were measured at both UCSC and Jet Propulsion Laboratory (JPL). The measurement results of electrical conductivity form 300 K to 600 K at the two different groups are shown in Figure 2.

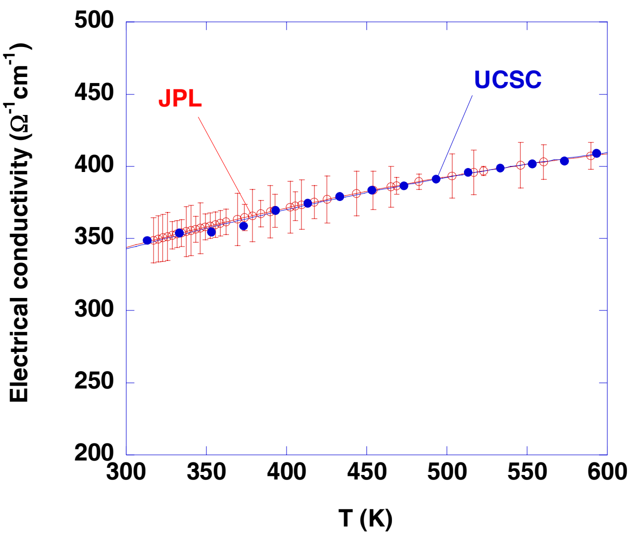

Figure 2. Electrical conductivity of $(InGaAs)_{0.8}(InAlAs)_{0.2}$ with 0.3% ErAs nanoparticles measured at UCSC and JPL, respectively.

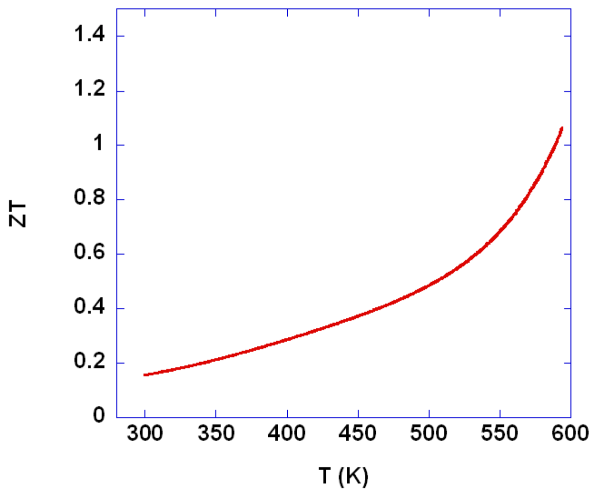

Figure 3. The ZT values of (InGaAs)0.8(InAlAs)0.2 with 0.3% ErAs nanoparticles based on the measurements on the thermal conductivity, electrical conductivity and Seebeck coefficients from temperature of 300 K up to 600 K.

Based on the measurements of thermal conductivity, electrical conductivity and Seebeck coefficient, the figure of merit of ErAs:InGaAlAs is known with the ZT value of 0.18 at 300 K, and greater than 1 at 600 K, as shown in Figure 3.

DEVICE FABRICATION

The segmented generator modules were fabricated via pickup and place approach method and flip-chip bonding techniques. Processing techniques similar to those of standard large scale integrated circuits were used for the fabrication of the segmented elements of ErAs:InGaAlAs and Bi_2Te_3.

MBE was used to grow epitaxial layers on two 75 mm InP wafers: one n-type of 50 µm ErAs: $(InGaAs)_{0.8}$ $(InAlAs)_{0.2}$ wafer and one p-type 50 µm of ErAs:InGaAs wafer.

The thin film element fabrication started with the front side metallization of the epitaxial layer: Ni/GeAu/Ni/Au contact metals were used for n-type ErAs:InGaAlAs, and Pt/Ti/Pt/Au were used for p-type ErAs:InGaAs, respectively. The InP substrate was removed through wet etching solution to expose the backside of the epitaxial layer. The backside metallization was similar to that of the front side: Ni/GeAu/Ni/Au and Pt/Ti/Pt/Au were used for n-type and p-type, respectively. Then the n and p type thin film wafers were diced into square chips ready for bonding.

Transmission line method (TLM) patterns were fabricated for the specific contact resistance measurements. The results show that both n and p type specific contact resistances were on the order of 10^{-7} $\Omega\cdot cm^2$.

Ni was used as contact metallization for Bi_2Te_3 of both n and p type. After metallization, the bulk Bi_2Te_3 was cut into square chips of 1 mm × 1 mm in area. All the Bi_2Te_3 elements were bonded on a lower ceramic plate; while the ErAs:InGaAlAs elements were bonded on an upper ceramic plate. Finally, the lower Bi_2Te_3 bonded plate and the upper ErAs:InGaAlAs bonded plate were bonded together using flip-chip bonding to form a 254 element generator module.

MEASUREMENT RESULTS AND DISCUSSIONS

The measurement setup which consists of a heat sink with water cooling, a heat source consisting of two cartridge heaters, thermal couples for temperature monitoring and electrical probes. Two thermocouples were used for temperature measurements: one of the thermocouples was on the top of the generator; the other was at the interface of the heat sink and the generator. The output current flowing through the power generator and the output voltage generated across an external electrical resistor load were measured. The data acquisition was done via four multimeters controlled by a computer.

The measurement results of output power are shown in Figure 4. When the heat source temperature rises above 500 K, the output power gradually shows some saturation. There are two reasons for this: i) the ZT value of Bi_2Te_3 dropped at high temperature; ii) the temperature at the cold side of the generator module also began to rise due to the high heat flux through the elements, so that the temperature difference across the element didn't increase linearly with the rise of the heat source temperature. The output power can be improved by improving the thermoelectric properties of the elements, reducing the thermal and electrical parasitic loss, and increasing heat transfer coefficient of the heat sink, which will effectively increase the temperature difference across the elements.

CONCLUSIONS

The incorporation of ErAs nanoparticles into $(InGaAs)_{1-x}(InAlAs)_x$ alloy results in significant improvement in the material's thermoelectric properties. Variable temperature measurements show that the ZT value reaches 1 around 600K, which is very encouraging. Segmented generator modules were fabricated using ErAs: InGaAlAs and Bi_2Te_3. Output power up to 5.5 W weremeasured with heat source temperature up to 600 K. The performance of thermoelectric generator modules can be further improved by improving material thermoelectric properties, and reducing electrical and thermal parasitic resistance loss, and improve the heat transfer coefficient of the heat sink.

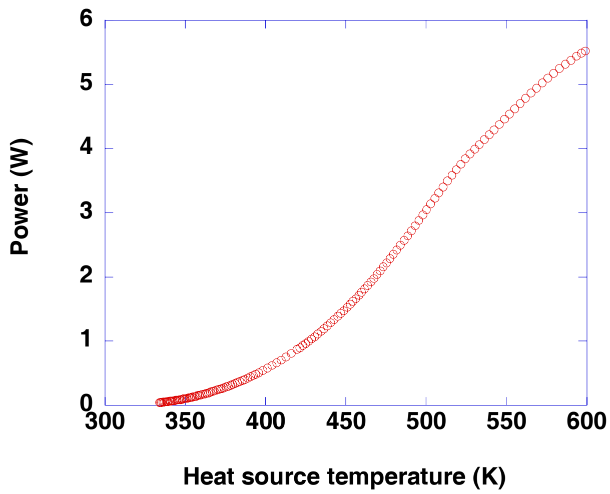

Figure 4. The output power measurement results for the 254 segment element power generator of 50 μm ErAs:$(InGaAs)_{1-x}(InAlAs)_x$ and 300 μm Bi_2Te_3.

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